LCA IN TRANSPORTATION

LCA of petroleum-based lubricants: state of art and inclusion of additives

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Abstract

Purpose While the application of Life Cycle Assessment (LCA) to lubricants can be considered fully operational for general purposes outside the lubricants industry, where Life Cycle Inventories (LCIs) of mineral and synthetic base oils can be used interchangeably and where additives can be excluded, this is not the case for research and development purposes within the industry. Previous LCAs of base oils are not sufficiently detailed and comprehensive for R&D purposes, and there are no LCAs of lube additives and fully formulated lubricants. The aim of this paper is to integrate and expand previous LCAs of base oils and to investigate on the contribution of lube additives to the environmental impacts of a fully formulated lubricant.

Materials and methods This study considers three base oils (mineral, poly-alpha-olefins (PAO) and hydrocracked) and a set of lubricating additives typically used in fully formulated engine oil. The LCA model is based on both industry and literature data.

Results and discussion Trends in the lubricants industry towards more sophisticated base oils correspond to remarkably higher environmental impacts per kilogram of product but lead to reduced impacts per kilometre. The contribution of additives to the life cycle impacts of commercial lube oil was found to be remarkably high for some impact categories (nearly 35 % for global warming).

Conclusions As base oil is concerned, this study made the point on data availability and provided a contribution in order to integrate and expand previous LCAs of mineral base oil and PAO. On the side of additives, the main conclusion is that in modern lubricants, the contribution of additives in terms of environmental impact can be remarkably high and, therefore, they should not be excluded.

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1 Introduction

Lubricants are important products of the refining industry (European Commission DG Energy 2010a). They are widely used to reduce friction between moving components and in modern engines. There are nearly unlimited applications for standardised or specifically tailored lubricants, and the related industry has evolved in time to meet the expectations of customers with a large variety of new products. The world demand in 2010 was 34.5 MT: 56 % automotive lubricants, 26 % industrial lubricants, 8 % greases and

10 % process oils. Moreover, it is estimated that from 5,000 to 10,000 different lubricants are necessary to satisfy more than 90 % of all applications (Mang and Dresel 2007).

Fully formulated lubricants are constituted by a base oil, typically coming from refining of crude oil, and additives that are used to enhance the performance of lubrication as well as to mitigate drawbacks such as corrosion and wear (Mortier and Orszulik 1997). Base oils can be roughly divided in two categories: mineral and synthetic.

Beyond the technological and economic aspects, the petrochemical sector, and thus the lubricants industry, is claimed to be responsible of heavy and well-perceived environmental impacts that have pushed companies to invest in clean technologies and more environmentally friendly products (Bevilacqua and Braglia 2002).

In such a context, Life Cycle Assessment (LCA) has been used for several years in the lubricants field in two directions: (1) understanding and reducing environmental impacts of lube products and (2) including Life Cycle Inventory (LCI) data of lubricants in LCAs of products and services. According to an analysis of the scientific literature on LCA of lubricants, most of applications refer to the second type, i.e., for purposes outside the lubricants industry, and are typically intended to create databases such as Boustead Model (2005), ELCD (2010) and Ecoinvent (2007) in order to support LCA practitioners in modelling thousands of processes where lubricants are directly or indirectly involved.

On the contrary, LCAs intended to be used as a tool to understand/improve petroleum-based lubricants is much more restricted, as it is highlighted by the limited number of dedicated scientific papers (Serra-Holm 2004; Vag et al. 2002; Wang et al. 2004). In a few other papers, LCA is used to compare bio-lubricants against generic (or not well-defined) mineral base oils (Ekman and Börjesson 2011; Miller et al. 2007). Moreover, in all of the above references, lube additives are excluded using either one of the following justifications: the quantity is low (Ecoinvent 2007; Ekman and Börjesson 2011) or, in comparative LCAs, the quantities are assumed to be similar in all products (Miller et al. 2007).

As lubricants are concerned, the state-of-art suggests that LCA can be considered fully (or almost) operational for general purposes outside the lubricants industry, where LCIs of mineral and synthetic base oils can be used interchangeably and where additives can be excluded.

On the contrary, given that in recent years there was a clear shift towards more sophisticated base oils, which are likely to correspond to higher environmental impacts per kilogram and there was a tendency to massively use conventional and innovative additives in several applications, it clearly emerges that LCA is far to be operational within the lubricants industry. This lack of detailed, updated and reliable LCIs is the situation faced by the partners of the EU-FP7 research project AddNano (http://sites.google.com/site/addnanoeu/), an 11-million-euro project that involves worldwide leading companies in the lubricating sector and which is targeted to the development and the scale-up of innovative fully formulated lubricating oils incorporating nano-particles. Advanced nano-materials have shown initial promising attitudes for reducing friction and enhancing protection against wear (Feldman et al. 1996; Feldman et al. 2000) and are presently under study. With focus on engine oil (crankcase) applications, among other technological objectives, the AddNano project is using LCA in order to understand and improve the environmental performance of innovative nano-based lubricants in a life cycle perspective.

Given that the new nano-components are intended to be used in substitution or in mix with conventional additives and that several base oils are to be tested, it clearly emerged that background LCI data were not sufficient and that an additional effort was required to the project partners in order to reasonably expand and complement the background dataset. This appeared to be a situation where the direct involvement of important industrial partners could partially fill the gap of data related to the LCA of additives. Such a data gap can partially be justified by the enormous variety of additives presently available and by the fact that additive producers are extremely conservative and seldom available to supply data and information that they consider strictly confidential.

Bearing this in mind, the focus and the novelty brought by this paper are twofold and can be summarised as follows. As a first step, a critical review of literature LCAs of base oils is carried out and used to create an updated from-cradle-to-gate model of mineral base oil and poly-alpha-olefins (PAO) base oil. The environmental impacts are compared with those relevant to the hydrocracked base oil in the Ecoinvent (2007), and the results are discussed and contrasted against the recent trends in the lubricants industry. As a second step, a simplified LCA of the most common lube additives is carried out using both data from the industry and from the literature. Although simplified, this is, in fact, an opportunity to start a process of cooperation with additive producers and an occasion to clarify whether the contribution of additives to the impacts of fully formulated lubricants is always negligible or not. In fact, Ekman and Börjesson (2011) exclude additives from their analysis, but they admit that in applications where the quantity of additives can be up to 30 % of fully formulated lubricants and in case of indicators related to human and/or eco-toxicity, the environmental consequences could be remarkable and, finally, they recommend to include additives in future LCAs.

2 Materials and methods

This study considers three base oils (mineral, PAO and hydrocracked) and a set of additives typically used in an

engine lube oil according to the average composition reported in Table 1. Mineral base oils are produced via refining the residual fraction of crude oil, while synthetic base oils usually are prepared through the reaction of chemical compounds, which are often petroleum-derived. Hydrocracking also represents an important process in the production of modern lube oils, mainly due to the low influence of the quality of crude oil on the quality of final products (Mang and Dresel 2007).

Lubricating additives constitute today an important fraction of a fully formulated oil and are necessary to meet the stringent requirements of modern engines, enhancing the lubricating properties of lube oils as well as enlarging and stabilising the range of operability under severe conditions of aging and temperatures (Rudnick 2009). The additives considered in this work are representative of the following categories: detergents, dispersants, viscosity modifiers, antioxidants and antiwear (see Table 1).

2.1 Data sources

As stated above, there are some literature references relevant to the LCA of base oils, but none about additives. The main data sources for the LCA presented in this paper are summarised in Table 2. Among these, there are databases such as Boustead Model (2005), Ecoinvent (2007), European Reference Life Cycle Database-ELCD (2010), the IFEU/GEIR report issued by the lubricants industry (Fehrenbach 2005) and the Reference Document on Best Available Techniques for Mineral Oil and Gas Refineries-BREF (European IPPC Bureau 2003). It must be said that such data sources have very different bases for their data, which can generate heavy inconsistencies. However, it must also be said that in all the references quoted in Table 2, the authors state that large datasets were not communicated by the industry and that heavy assumptions or exclusions were undertaken. For that reason, the close cooperation with the AddNano partners (PETRONAS) was extremely helpful to identify the most suitable sources in different steps of LCA modelling.

Table 1 Typical composition of an engine lube oil (source: personal communication from PETRONAS)

Component	Percentage (%)
Base oil (mineral/synthetic)	80
Detergents	2
Dispersants	6
Viscosity modifiers (OCP—olefins copolymers)	9
Antioxidant (ZDDP—zinc dialkyldithiophosphates/phenols)	1
Antiwear (ZDDP—zinc dialkyldithiophosphates)	2

Table 2 Main sources containing data/information on LCI of lubricants

Source	Title	Year of publication	Main content	Allocation criteria	Inclusion of additives and/or remarks
Boustead (2005)	Primary oil refining	2005	LCA of lube oil (unspecified)	Energy	No
Ecoinvent (2007)	Lubricants	2007	LCI of lube oil (hydrocracked)	Mass	No/impact of additives is considered negligible
ELCD (EC, JRC, IES, 2010)	EU15—lubricants from crude oil, consumption mix, at refinery	2010	LCI of lube oil (unspecified)	Energy consumption (thermal energy, steam and electricity) of processes is allocated by mass. Raw materials (crude oil) consumption is allocated by energy	No
IFEU/GEIR (Fehrenbach 2005)	Ecological and energetic assessment of re-refining used oils to base oils: substitution of primarily produced base oils including semi-synthetic and synthetic compounds (commissioned by GEIR—Groupement Européen de l'Industrie de la Régénération)	2005	LCA of five regeneration techniques for re-refining used oil. Comparison of re-refined oils with a mineral base oil and a PAO base oil (synthetic)	Mass	No
BREF (European IPPC Bureau 2003)	Reference document on best available techniques for mineral oil and gas refineries	2003	Data on average raw materials consumption and emissions of oil refineries	None	No

The LCI datasets for mineral base oil and synthetic PAO are reported as Electronic Supplementary Material. The layout of flowcharts is mostly based on the IFEU/GEIR report (Fehrenbach 2005). As mineral base oil is concerned, energy and raw materials consumption during refining were mostly retrieved using average data from BREF (European IPPC Bureau 2003), and the impacts connected with the extraction and transport of crude oil were adjusted using data from DG Energy of the EC (2010b). PAO base oil was mostly modelled based on IFEU/GEIR data. The hydro-cracked base oil included in the Ecoinvent (2007) was used as term of comparison (LCI data are not reported in this paper). The simplified LCA of additives was developed using personal communication from the AddNano partners (PETRONAS and Centro Ricerche FIAT) and literature data that will be discussed in "Section 3.3".

2.2 System boundaries and functional unit

The LCA models here proposed cover the phases of extraction, transportation and production until the exit of the refinery/factory in a from-cradle-to-gate perspective. The production process has been divided in sub-units (flow charts are reported as Electronic Supplementary Material). Each sub-unit represents a single step necessary to transform the raw materials into base oil. In each step, raw materials and energy consumption are considered as input and products and co-products as output as well as inputs to the successive process. The analysis has been set on European average data, representative of the European refinery industries.

Given the main purpose of the present study, i.e., discussing on availability and comprehensiveness of LCI data for future research and development within the lubricants industry, the principal functional unit is 1 kg of final product. However, in the light of the recent trends in the lubricants industry and according to industry data communicated by the AddNano partners, the lifetime of engine oil in crankcase applications increased from an average of 15,000 km (mineral base oil) to 25,000 km (PAO and hydrocracked), whereas the quantity utilised in a middle-sized diesel engine remained unchanged (4 kg). Accordingly, the results will be presented per kilogram of product and per kilogram adjusted to the lifetime (kilometre-adjusted).

2.3 Allocation criteria

Due to the fact that refineries are highly integrated and produce multiple outputs, this determines the need to define a criterion of allocation to distribute the environmental burdens to each product. The allocation can be done considering different parameters: mass, energy content and market price (Ekman and Börjesson 2011; Wang et al. 2004).

In this study, the allocation criterion for outputs is mass. This was considered the most appropriate in the context of the AddNano project, which is more focused on additives and fully formulated lubricants than on base oils. The consequences of a different choice on allocation are discussed by Ekman and Börjesson (2011) and in Wang et al. (2004).

As far as allocation of input data is concerned, according to the BREF report (European IPPC Bureau 2003), most air emissions are related to the use of energy. It is, therefore, possible to allocate input energy and materials according to the activities highlighted in the flowcharts reported as Electronic Supplementary Material. Allocation of water emissions and generated waste between base oil and other refinery products was carried out at refinery level (see Electronic Supplementary Material).

2.4 Selection of the environmental impact indicators

The Life Cycle Impact Assessment method used in order to show the results of this study is the IMPACT 2002+ (Jolliet et al. 2003). This method is composed of 15 midpoint indicators as it will be shown in "Section 4".

3 System description

Mineral base oil and synthetic base (PAO) are investigated first. The description turns, then, on lubricating additives.

3.1 Mineral base oil

Mineral base oil is produced from crude oil through several processes of distillation and refinery. Detailed process data were retrieved from the IFEU Report (Fehrenbach 2005). Average values of energy and materials consumption are calculated on the basis of the BREF (European IPPC Bureau 2003).

A crude oil mix coming from different countries has been considered, based on the European Commission energy average data on oil imports and deliveries (European Commission DG Energy 2010b). The corresponding areas of extraction reported by the European bureau have been associated to the available units in the Ecoinvent database (see Electronic Supplementary Material). The Ecoinvent considers both the impacts related to the phase of extraction and to the transport from on-shore or off-shore sites to the refinery (transportation system: oil pipeline and transoceanic tanker).

The production flow chart considered in the study is shown in Electronic Supplementary Material (F1). The basic distillations (atmospheric and vacuum) are the first steps of the process, aimed to separate the base oil feedstock from the other petroleum products. Afterwards, the waxy distillate gets through specific refining stages that purify the base oil from unwanted components.

Accordingly, 112 kg of base oil are produced from 1 t of crude oil. Waxy distillate represents around 20 % of the distillation products and only half of it is turned into base oil. For each step of the process, the energy consumption and materials inputted as well as the quantity of output products and co-products are reported in the Electronic Supplementary Material (F1).

3.2 Synthetic base oil (PAO)

There are many kind of base oil stocks for synthetic lubricants such as PAO, alkylated aromatics, polybutenes, etc. In this study, PAO are considered as representative of synthetic bases.

The term PAO refers to hydrogenated oligomers of α-olefin, usually α-decene. PAO are synthesised from 1-decene monomers to branched trimmers of 30 carbon atoms; 1-decene is gained from a fraction of linear α-olefins (LAO) which is polymerised from ethylene molecules (Mang and Dresel 2007; Mortier and Orszulik 1997).

Data relevant to energy and materials consumption and emissions were retrieved from the IFEU/GEIR Report (Fehrenbach 2005). The production chain has been modelled considering processes and relative intermediate products shown in the Electronic Supplementary Material (F2). In particular, the feedstock input is composed by naphta from crude oil distillation (around 60 %) and by gas condensate from natural gas processing; 10 % of feedstock is converted in PAO through three stages of synthesis (steam cracking, LAO and PAO synthesis).

3.3 Lubricating additives

The lubricating additives industry is continuously evolving in order to improve properties and performances of commercial lubricants, reduce fuel consumptions and increase the lifetime of engines. According to experts, there are still large margins for improvement in the development of new additives that can enhance lubricant performances and reduce environmental burdens (Rudnick 2009).

Due to the absence of specific literature and the low availability of primary data, a simplified methodology has been adopted to carry out a LCA of lube additives in cooperation with the AddNano project partners.

The following steps were undertaken:

1. Identification of the additive categories typically used in average engine lube oil. The chemical composition of a conventional engine oil has been identified using personal communication from PETRONAS and literature data (Mang and Dresel 2007; Rudnick 2009).
2. Selection of a representative additive for each category.
3. Identification of the correspondence between the selected additive and an industrial product available in the Ecoinvent database (2007): *proxy product criterion*.
4. When no straightforward correspondence between additives and Ecoinvent units could be found, a secondary criterion was adopted: *proxy synthesis process criterion*. The correspondence with one or more Ecoinvent entries has been searched through a simplified chemical composition and according to a proxy synthesis process.

Based on the average composition of engine lube oil reported in Table 1, the main data and assumptions for the simplified LCA of additives are reported in Table 3. A short description of additives and a few comments on their function in fully formulated lubricants are reported in the following paragraphs.

3.3.1 Detergents

Detergents represent an important class of the so-called overbased additives, which are colloidal particles of calcium carbonate and hydroxide, stabilised by a surfactant layer.

Table 3 Main data and assumptions for the simplified LCA of additives

Category	Selected additive	Proposed correspondence (database Ecoinvent)	Relative composition	Decision criteria
Detergents	Alkylbenzenesulfonic acid	Alkylbenzene sulphonate, linear, petrochemical at plant	100 %	Proxy product
Dispersants	Polyisobutenyl succinimide	Synthetic rubber at plant	100 %	Proxy product
Viscosity modifiers (OCP)	Olefin co-polymers	*n*-Olefins at plant	100 %	Proxy product
Antioxidant (ZDDP/phenols)	Phenolic antioxidants	Phenols at plant	50 %	Proxy product
	Zinc dialkyldithiophosphates (ZDDP)	Isobutanol at plant	29 %	Proxy synthesis process
		Zinc oxide at plant	7 %	
		Hydrogen sulphide (H_2S) at plant	7 %	
		Phosphorus chloride at plant	7 %	
Antiwear (ZDDP)	Zinc dialkyldithiophosphates (ZDDP)	Isobutanol at plant	58 %	Proxy synthesis process
		Zinc oxide at plant	14 %	
		Hydrogen sulphide (H_2S) at plant	14 %	
		Phosphorus chloride at plant	14 %	

They are formed by a long chain of polar hydrophilic hydrocarbons and a polar hydrophilic head. The oleophilic hydro serves as solubiliser in the base fluid, while the head attracts contaminants within the lubricant. The detergents are metal-containing and the most diffused are the sulphonates, followed by phenates, salicylates and phosphonates (Hudson et al. 2006; Rudnick 2009).

3.3.2 Dispersants

The role of dispersants is to prevent agglomeration of particles produced by oil degradation and metallic parts wear (sludge) and maintain them in suspension in the oil. Even if the principle of operation is similar to detergents, they differ from detergents because they are, by definition, free of metals. The most diffused dispersant are polybutenes and, in particular, the polyisobutenylsuccinimide type (Hui et al. 1997).

3.3.3 Viscosity modifiers

Viscosity is a fundamental characteristic of lubricating fluids in order to operate correctly mechanical moving parts. Pressure and temperatures are the parameters that influence more the lube oil. Viscosity modifiers aim at optimising the working efficiency.

They are constituted by high molecular polymers, with a flexible molecular chain structure. Examples of viscosity modifiers are polymetacrylates (PMAs), polyethylenecopropylenes or the so-called olefin copolymers (OCPs) (Souza de Carvalho et al. 2010; Rudnick 2009).

3.3.4 Antioxidant

Antioxidants play an important role in lubricating oil to prevent processes of ageing that can deteriorate the quality of the lubrication. Aged lubricants can be typically characterised by common aspects such as discoloration or burnt odour (Rudnick 2009).

Methylene bridge hindered phenolic antioxidants, and alkylated diphenylamine antioxidants have demonstrated high performance to this purpose. These antioxidants are prepared by alkylation reactions, resulting in the formation of complex product mixtures (Greene and Gatto 1999).

3.3.5 Antiwear

The wear between sliding surfaces is an inevitable drawback of machines during start-up, running-in and transient operation. Antiwear additives in modern engines have to control wear at acceptable levels. Zinc dialkyldithiophosphates (ZDDPs) have been extensively used as antiwear additive since 1940s. They work with the principle of "boundary lubrication", protecting the moving parts against wear due to the formation of tribochemical films on the surfaces in contact (Barnes et al. 2001; Lin and So 2004; Varlot et al. 2001).

4 Results and discussion

Results related to the LCA of base oils are presented and discussed first. The discussion turns, then, on additives.
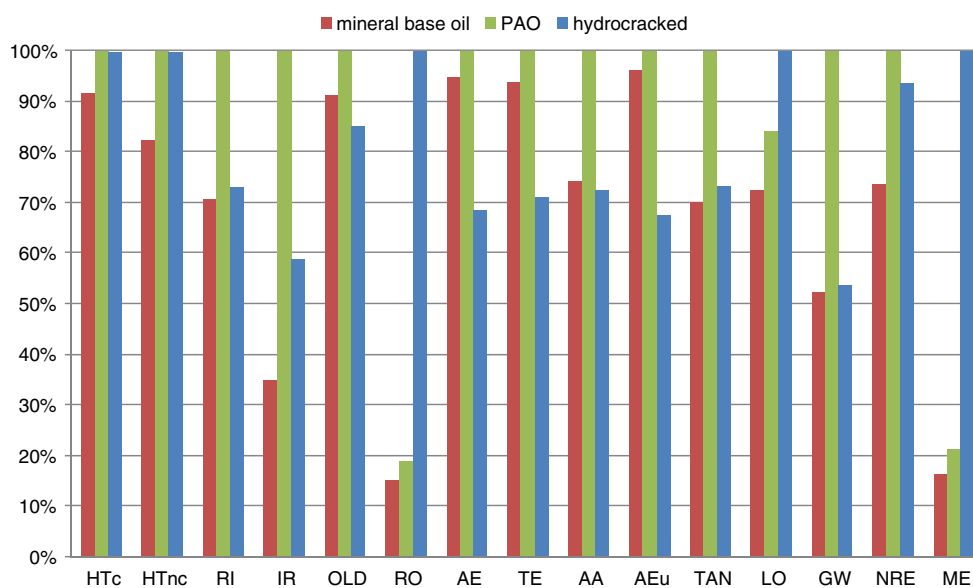
4.1 Base oils

Midpoint impact indicators obtained in this study for mineral base oil and PAO are reported in Table 4. Figure 1 shows a graphical comparison between mineral and PAO

Table 4 Environmental impacts in the production of mineral base oil and PAO (data per 1 kg)

Midpoint indicators		Unit	Mineral base oil	PAO
Human toxicity (carcinogens)	HT_c	kg C_2H_3Cl eq.	0.012	0.014
Human toxicity (non-carcinogens)	HT_{nc}	kg C_2H_3Cl eq.	0.013	0.015
Respiratory inorganics	RI	kg PM2.5 eq.	0.001	0.002
Ionizing radiation	IR	Bq C-14 eq.	16.00	45.89
Ozone layer depletion	OLD	kg CFC-11 eq.	7.0E-07	7.6E-07
Respiratory organics	RO	kg C_2H_4 eq.	0.001	0.001
Aquatic eco-toxicity	AE	kg TEG water	286.8	302.4
Terrestrial eco-toxicity	TE	kg TEG soil	61.64	65.70
Aquatic acidification	AA	kg SO_2 eq.	0.009	0.012
Aquatic eutrophication	AEu	kg PO_4 P-lim.	0.001	0.001
Terrestrial acidification/nitrification	TAN	kg SO_2 eq.	0.022	0.031
Land occupation	LO	m^2 org. arable	0.007	0.009
Global warming	GW	kg CO_2 eq.	0.985	1.880
Non-renewable energy	NRE	MJ primary	62.91	85.42
Mineral extraction	ME	MJ surplus	0.004	0.005

Fig. 1 Comparison of from-cradle-to-gate environmental impacts related to mineral base oil, PAO base oil and base oil hydrocracked (data per 1 kg)

base oils and also includes impacts for the hydrocracked base oil according to the Ecoinvent database (the highest impacts are made equal to 100 %).

PAO shows the highest impacts in all the categories, with the exception of respiratory organics, land occupation and mineral extraction where the highest impacts are those of hydrocracked base oil. It can be observed that greenhouse emissions of PAO are almost twice than those of mineral base oil due to higher quantities of refinery gas burned for heat consumption and, in general, to a more energy-consuming production process.

These results show that trends in the lubricants industry towards more sophisticated base oils correspond to remarkably higher environmental impacts associated to 1 kg of product. This aspect could be explained by the fact that modern lubricants are produced by more complex and

energy-consuming processes. However, it has to be taken in mind that modern lubricating oils remarkably increase the lifetime of engine oil and, consequently, the mileage that can be covered.

As shown in Fig. 2, where the impacts are adjusted to the mileage covered in the engine oil lifetime, the reduced number of oil changes (from 15,000 km to 25,000 km) corresponds to reduced impacts per kilometre, i.e., a reduction in total impacts in a life cycle vision. Mineral base oil shows the highest impacts in 11 out of 15 indicators. However, it must be remarked that for some applications, engine oil lifetime can reach higher mileages (40,000 km) and that there are other aspects such as increased vehicle fuel economy and uncertainty of input data and LCA results that need further investigation and close cooperation with the industry.

Fig. 2 Comparison of from-cradle-to-gate environmental impacts related to mineral base oil, PAO base oil and base oil hydrocracked (data per 1 kg adjusted to the lubricant lifetime)
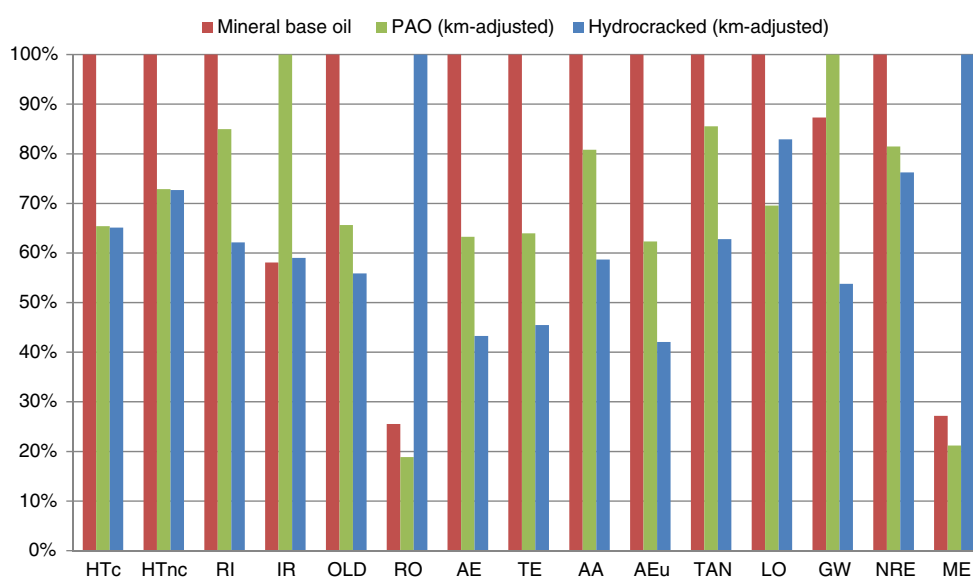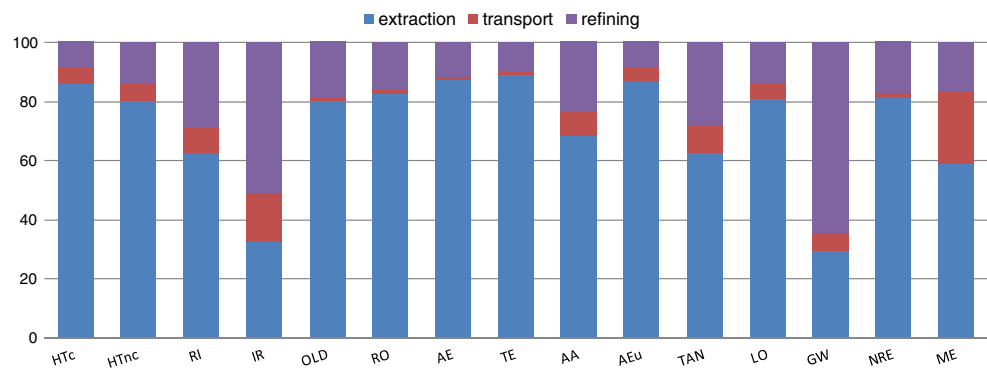
Fig. 3 Contribution analysis of the main phases in the production of mineral base oil

Figures 3 and 4 show the contribution analysis of the impacts attributable to mineral base oil and PAO. The phases considered are extraction of crude oil and natural gas, transport to the plant and base oil processing.

It is possible to observe that in case of mineral base oil, the phase of extraction shows the highest contribution, while the second position is generally held by refining. Exceptions are represented by ionising radiations and greenhouse emissions. The contribution of transport is always the lowest, but it is more than 20 % in case of mineral extraction. As PAO is concerned, the phase of refining appears to be remarkably more impacting, counting up to 90 %. Transport shows again the lowest contribution, always less than 10 %.

4.2 Lube additives in fully formulated engine oil

As previously said, fully formulated oils are composed by base oil and additives. Given the composition reported in Table 1 where the base oil is assumed to be mineral, the results of the life cycle analysis of fully formulated lube oil are reported in Fig. 5. The first column shows the composition in mass, so that it is possible to visually identify those components that give a contribution to the overall environmental impacts higher than the contribution in mass.

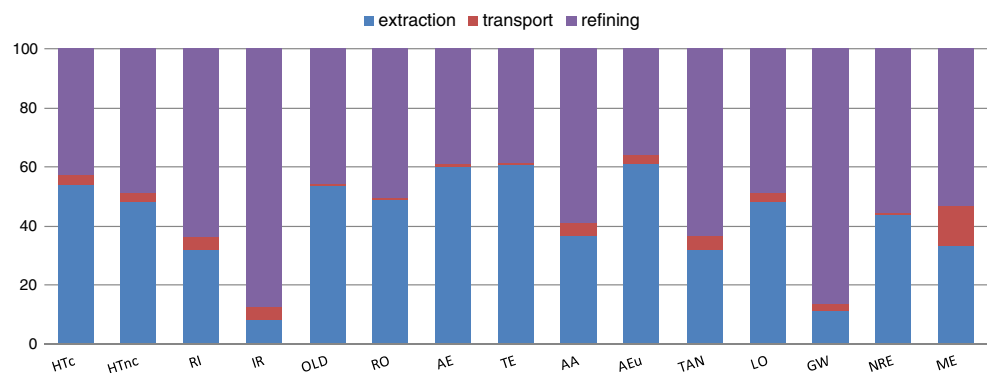It can be highlighted that the contribution of additives to the life cycle impacts of commercial lube oil cannot be considered negligible, as they can be up to 70 % of the total impacts, while they represent only 20 % in mass. In particular, for carcinogens and mineral extraction, the contribution of additives is more than 50 %.

These findings confirm and expand the statement of Ekman and Börjesson (2011) according to which additives should be explicitly considered in LCAs of lubricants. Moreover, the percentage of additives is increasing in time and gaining importance in modern lubricants. Therefore, if the contribution of lubricating additives to the environmental impacts could be considered negligible in previous LCAs, this simplification is not anymore valid for modern lubricants.

Mid-point impact indicators are typically regarded as fairly objective by LCA practitioners; therefore, they can be considered as background information for further LCA studies and discussion. However, it must be said that not all the 15 mid-point indicators are equally meaningful to the comparison between the three base oils. For instance, land occupation is likely to correspond to a lower concern than non-renewable energy or global warming.

With that in mind, the midpoint indicators of IMPACT 2002+ were normalised to the per capita yearly impacts of one European citizen, thus expressing the results as person-year equivalents. This is in order to further investigate on the relative importance of the obtained impacts. According to Fig. 6, the contribution of additives to the three highest normalised indicators is 25 % of respiratory inorganics, 35 % of greenhouse emissions and 24 % of non-renewable energy.

Fig. 4 Contribution analysis of the main phases in the production of PAO base oil
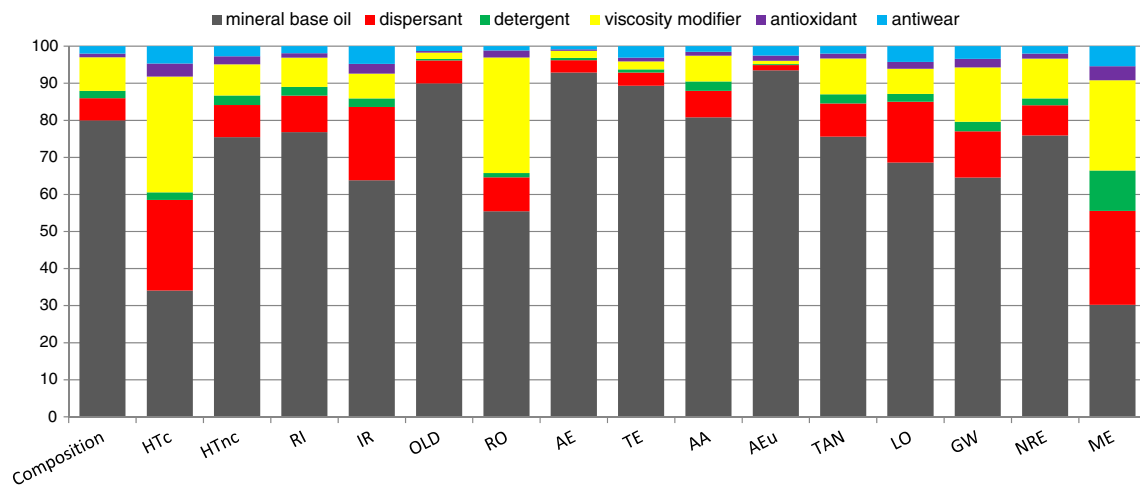
Fig. 5 Environmental impacts of fully formulated engine oil (characterisation step)

5 Conclusions

The results presented in this paper suggest that the application of LCA to lubricants cannot always be considered fully operational. If, for general purposes outside the lubricants industry, the exclusion of additives can be tolerated, this is not the case for research and development purposes where LCA of lubricants is still far to be operational. More detailed and comprehensive LCAs of different base oils are necessary and lube additives must be included in the LCA of modern fully formulated lubricants.

As far as LCA of base oil is concerned, this study made the point on data availability and provided a contribution in order to integrate and expand previous LCAs of mineral base oil and PAO. This was useful to highlight how modern synthetic lubricating base oils have higher impacts per kilogram in comparison to traditional mineral base oil. However, in reason of the fact that modern engines require lubricating oils that can lead to higher performance, reducing frictions and

fuel consumption, this can lead to environmental benefits in a life cycle perspective. Synthetic base oils offer a longer life time and require less oil changes, leading to a decrease of environmental impacts per distance covered. However, these overall environmental gains, although preliminarily quantified in this study, need further research and close cooperation with the industry.

On the side of additives and fully formulated lubricants, the main conclusion of this research is that in modern lubricants, the contribution of additives in terms of environmental impact can be remarkably high and, therefore, they cannot be excluded. Although simplified, this LCA of additives could represent a first step of a desirable cooperation with the additives industry, which so far kept information and data on processes and products strictly confidential. Moreover, it was highlighted that there is room for improvement in the production of additives and fully formulated lubricants through the deployment of new technologies such as those proposed in the AddNano project.
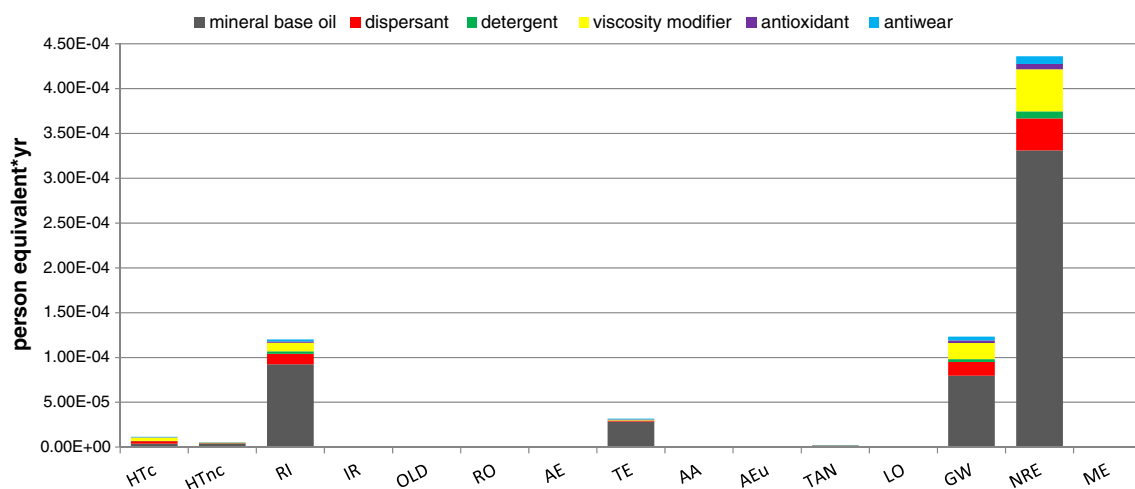

Fig. 6 Environmental impacts of fully formulated engine oil (normalisation step)

References

Barnes AM, Bartle KD, Thibon VRA (2001) A review of zinc dialkyldithiophosphates (ZDDPS): characterisation and role in the lubricating oil. Tribol Int 34(6):389–395

Bevilacqua M, Braglia M (2002) Environmental efficiency analysis for ENI oil refineries. J Clean Prod 10(1):85–92

Boustead Model 5.0 (2005) Operating manual. Boustead Consulting Ltd., HORSHAM, West Sussex, UK

EC, JRC, IES (2010) ELCD core database version II. European Commission, Joint Research Centre, Institute for Environment and Sustainability, Ispra (VA), Italy. Available at http://lct.jrc.ec.europa.eu/assessment/data

Ecoinvent (2007) Life cycle inventories of chemicals—Ecoinvent report no. 8. Swiss Centre for Life Cycle Inventories, Zürich and Dübendorf

Ekman A, Börjesson P (2011) Life cycle assessment of mineral oil-based and vegetable oil-based hydraulic fluids including comparison of biocatalytic and conventional production methods. Int J Life Cycle Assess 16(4):297–305

European Commission DG Energy (2010a) Commission staff working paper on refining and the supply of petroleum products in the EU.

European Commission DG Energy (2010b) Registration of crude oil imports and deliveries in the European Union (EU27)

European IPPC Bureau (2003) BREF—reference document on best available techniques for mineral oil and gas refineries. European Commission, Brussels

Fehrenbach H (2005) Ecological and energetic assessment of re-refining used oils to base oils: substitution of primarily produced base oils including semi-syntheticand synthetic compounds. IFEU Ltd. (commisioned by GEIR - Groupement Européen de l'Industrie de la Régénération), Heidelberg

Feldman Y, Frey GL, Homyonfer M, Lyakhovitskaya V, Margulis L, Cohen H, Hodes G, Hutchison JL, Tenne R (1996) Bulk synthesis of inorganic fullerene-like MS2 (M=Mo, W) from the respective trioxides and the reaction mechanism. J Am Chem Soc 118 (23):5362–5367

Feldman Y, Zak A, Popovitz-Biro R, Tenne R (2000) New reactor for production of tungsten disulfide hollow onion-like (inorganic fullerene-like) nanoparticles. Solid State Sci 2(6):663–672

Greene SV, Gatto VJ (1999) Size-exclusion chromatography method for characterizing low-molecular-mass antioxidant lubricant additives. J Chromatogr A 841(1):45–54

Hudson LK, Eastoe J, Dowding PJ (2006) Nanotechnology in action: overbased nanodetergents as lubricant oil additives. Adv Colloid Interface Sci 123–126:425–431

Hui F, Machtalère G, Xie J, Kolodziejczyk H, Rosset R (1997) Isolation of polybutenylsuccinimide-type dispersants from multi-grade lubricating oils by classical cation exchange chromatography. Anal Chim Acta 339(1–2):109–113

ELCD core database version II (2010).

Jolliet O, Margni M, Charles R, Humbert S, Payet J, Rebitzer G, Rosenbaum R (2003) IMPACT 2002+: a new life cycle impact assessment methodology. Int J Life Cycle Assess 8(6):324–330

Lin YC, So H (2004) Limitations on use of ZDDP as an antiwear additive in boundary lubrication. Tribol Int 37(1):25–33

Mang T, Dresel W (2007) Lubricants and lubrication. Wiley, Weinheim

Miller SA, Landis AE, Theis TL, Reich RA (2007) A comparative life cycle assessment of petroleum and soybean-based lubricants. Environ Sci Technol 41(11):4143–4149

Mortier R, Orszulik S (1997) Chemistry and technology of lubricants. Chapman & Hall, London

Rudnick L (2009) Lubricant additives: chemistry and applications. CRC, Boca Raton

Serra-Holm V (2004) Life cycle assessment (LCA) from cradle to gate of base oils used in the production of lubricants. In: Technische Akademie Esslingen international tribology colloquium proceedings, 2004. pp 549–552

Souza de Carvalho MJ, Rudolf Seidl P, Pereira Belchior CR, Ricardo Sodré J (2010) Lubricant viscosity and viscosity improver additive effects on diesel fuel economy. Tribol Int 43(12):2298–2302

Vag C, Marby A, Kopp M, Furberg L, Norrby T (2002) A comparative life cycle assessment of the manufacture of base fluids for lubricants. J Synthetic Lubric 19(1):39–57

Varlot K, Kasrai M, Bancroft GM, Yamaguchi ES, Ryason PR, Igarashi J (2001) X-ray absorption study of antiwear films generated from ZDDP and borate micelles. Wear 249(12):1029–1035

Wang M, Lee H, Molburg J (2004) Allocation of energy use in petroleum refineries to petroleum products. Int J Life Cycle Assess 9(1):34–44